Seminar on the optical properties of semiconductors

Free carrier transitions

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Optical dipole transitions

Crystal eigenstates:

$$H_0 = \hbar \sum_{\lambda \mathbf{k}} \epsilon_{\lambda,\mathbf{k}} |\lambda \mathbf{k}\rangle\langle\lambda \mathbf{k}| \qquad H_0 = \frac{p^2}{2m_0} + V_0(\mathbf{r})$$

Interaction with light field:

$$H_I = -d\,\mathcal{E}(t) = -er\,\mathcal{E}(t) =$$

$$-e\mathcal{E}(t) \sum_{\mathbf{k},\mathbf{k}',\lambda,\lambda'} r_{\lambda,\lambda'}(\mathbf{k},\mathbf{k}')|\lambda'\mathbf{k}'\rangle\langle\lambda\mathbf{k}|$$

$$r_{\lambda'\lambda}(\mathbf{k}',\mathbf{k}) = \langle\lambda'\mathbf{k}'|r|\lambda\mathbf{k}\rangle$$

$$\mathbf{r}_{\lambda,\lambda'}(\mathbf{k},\mathbf{k}') = \frac{\langle\lambda'\mathbf{k}'|[\mathbf{r},H_0]|\lambda\mathbf{k}\rangle}{\hbar(\epsilon_{\lambda,\mathbf{k}} - \epsilon_{\lambda',\mathbf{k}'})} = \frac{i\langle\lambda'\mathbf{k}'|\mathbf{p}|\lambda\mathbf{k}\rangle}{m_0(\epsilon_{\lambda,\mathbf{k}} - \epsilon_{\lambda',\mathbf{k}'})}$$

Matrix element of momentum operator

$$\langle \lambda' \mathbf{k}' | \mathbf{p} | \lambda \mathbf{k} \rangle = \int_{L^3} d^3 r \, \psi^*_{\lambda'}(\mathbf{k}', \mathbf{r}) \mathbf{p} \psi_\lambda(\mathbf{k}, \mathbf{r}) \simeq$$

$$\psi_\lambda(\mathbf{k}, \mathbf{r}) \simeq e^{i\mathbf{k}\cdot\mathbf{r}} \frac{u_\lambda(0, \mathbf{r})}{L^{3/2}}$$

$$\frac{1}{L^3} \int_{L^3} d^3 r \, e^{-i(\mathbf{k}' - \mathbf{k})\cdot\mathbf{r}} u^*_{\lambda'}(0, \mathbf{r})(\hbar \mathbf{k} + \mathbf{p}) u_\lambda(0, \mathbf{r}) \simeq$$

$$\sum_{n=1}^{N} \frac{e^{-i(\mathbf{k}' - \mathbf{k})\cdot\mathbf{R}_n}}{N} \int_{l^3} d^3 r \, \frac{e^{-i(\mathbf{k}' - \mathbf{k})\cdot\mathbf{r}}}{l^3} u^*_{\lambda'}(0, \mathbf{r})(\hbar \mathbf{k} + \mathbf{p}) u_\lambda(0, \mathbf{r})$$

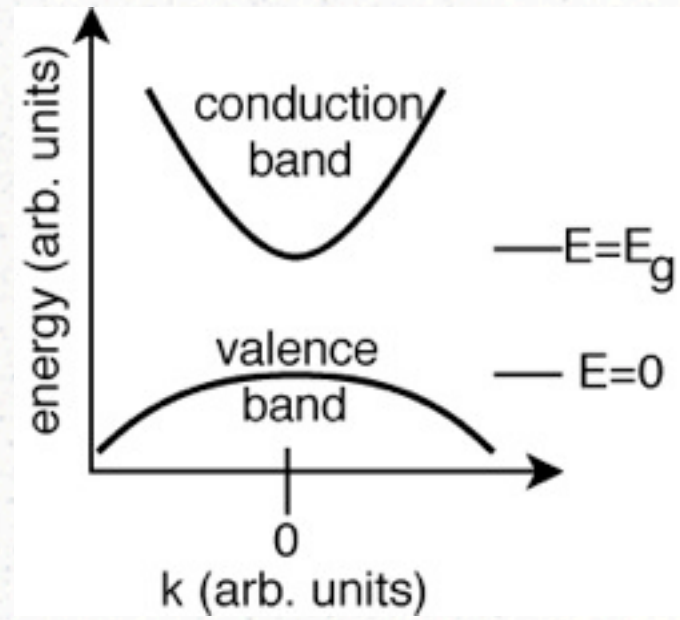
Optical dipole matrix element

$$\langle \lambda' \mathbf{k}' | \mathbf{p} | \lambda \mathbf{k} \rangle = \frac{\delta_{\mathbf{k},\mathbf{k}'}}{l^3} \int_{l^3} d^3 r \, u^*_{\lambda'}(0,\mathbf{r}) \mathbf{p} \, u_\lambda(0,\mathbf{r}) \equiv \delta_{\mathbf{k},\mathbf{k}'} \mathbf{p}_{\lambda',\lambda}(0)$$

$$\mathbf{d}_{\lambda'\lambda}(\mathbf{k}',\mathbf{k}) = e\mathbf{r}_{\lambda'\lambda}(\mathbf{k}',\mathbf{k}) = \frac{ie}{m_0(\epsilon_{\lambda',\mathbf{k}} - \epsilon_{\lambda,\mathbf{k}})} \delta_{\mathbf{k},\mathbf{k}'} \mathbf{p}_{\lambda'\lambda}(0)$$

$$\mathbf{d}_{\lambda'\lambda}(\mathbf{k}',\mathbf{k}) = \delta_{\mathbf{k},\mathbf{k}'} \mathbf{d}_{\lambda'\lambda}(0) \frac{\epsilon_{\lambda',0} - \epsilon_{\lambda,0}}{\epsilon_{\lambda',\mathbf{k}} - \epsilon_{\lambda,\mathbf{k}}}$$

$$\mathbf{d}_{\lambda'\lambda}(0) = \frac{ie\mathbf{p}_{\lambda'\lambda}(0)}{m_0(\epsilon_{\lambda',0} - \epsilon_{\lambda,0})}$$



$$\hbar\epsilon_{\lambda',\mathbf{k}} = E_g + \frac{\hbar^2 k^2}{2m_{\lambda'}}$$

$$\hbar\epsilon_{\lambda,\mathbf{k}} = \frac{\hbar^2 k^2}{2m_\lambda}$$

$$\mathbf{d}_{\lambda'\lambda}(\mathbf{k}',\mathbf{k}) = \delta_{\mathbf{k},\mathbf{k}'} \mathbf{d}_{\lambda'\lambda}(0) \frac{E_g}{E_g + \frac{\hbar^2 k^2}{2}\left(\frac{1}{m_\lambda} + \frac{1}{m_{\lambda'}}\right)}$$

Kinetics of optical interband transitions

Two-band model: $\lambda = c,\, v$

$$\mathcal{H}_I = -\mathcal{E}(t) \sum_{\mathbf{k},\{\lambda\neq\lambda'\}=\{c,v\}} d_{\lambda'\lambda}|\lambda'\mathbf{k}\rangle\langle\lambda\mathbf{k}| \equiv \sum_{\mathbf{k}} \mathcal{H}_{I,\mathbf{k}} \qquad \mathcal{H}_{I,\mathbf{k}} = -\mathcal{E}(t)(\, d_{cv}\,|c\mathbf{k}\rangle\langle v\mathbf{k}| + d^*_{cv}|v\mathbf{k}\rangle\langle c\mathbf{k}|)$$

$$\mathcal{H}^{int}_{I,\mathbf{k}}(t) = \exp\left(\frac{i}{\hbar}\mathcal{H}_0 t\right)\mathcal{H}_{I,\mathbf{k}}\exp\left(-\frac{i}{\hbar}\mathcal{H}_0 t\right) = -\mathcal{E}(t)\left[e^{i(\epsilon_{c,\mathbf{k}}-\epsilon_{v,\mathbf{k}})t} d_{cv}|c\mathbf{k}\rangle\langle v\mathbf{k}| + h.c.\right]$$

Single particle density matrix for the state k: $\boldsymbol{\rho_{\mathbf{k}} = \sum_{\lambda',\lambda} \rho_{\lambda',\lambda}(\mathbf{k},t)|\lambda'\mathbf{k}\rangle\langle\lambda\mathbf{k}|}$

$$\rho^{int}_{\mathbf{k}}(t) = \exp\left(\frac{i}{\hbar}\mathcal{H}_0 t\right)\rho_{\mathbf{k}}(t)\exp\left(-\frac{i}{\hbar}\mathcal{H}_0 t\right)$$

Equation of motion for the density matrix:

$$\frac{d}{dt}\rho_{\mathbf{k}}(t) = -\frac{i}{\hbar}[\mathcal{H}_{\mathbf{k}}\ ,\ \rho_{\mathbf{k}}(t)] \qquad \frac{d}{dt}\rho^{int}_{\mathbf{k}}(t) = -\frac{i}{\hbar}[\mathcal{H}^{int}_{I,\mathbf{k}}\ ,\ \rho^{int}_{\mathbf{k}}(t)]$$

$$\frac{d}{dt}\rho^{int}_{\mathbf{k}}(t) = \frac{i}{\hbar}\mathcal{E}(t)\sum_{\lambda',\lambda}\rho^{int}_{\lambda'\lambda}(\mathbf{k},t)$$

$$\times\Big[e^{i(\epsilon_{c,\mathbf{k}}-\epsilon_{v,\mathbf{k}})t} d_{cv}\left(|c\mathbf{k}\rangle\langle v\mathbf{k}|\lambda'\mathbf{k}\rangle\langle\lambda\mathbf{k}| - |\lambda'\mathbf{k}\rangle\langle\lambda\mathbf{k}|c\mathbf{k}\rangle\langle v\mathbf{k}|\right)$$

$$+e^{-i(\epsilon_{c,\mathbf{k}}-\epsilon_{v,\mathbf{k}})t} d^*_{cv}\left(|v\mathbf{k}\rangle\langle c\mathbf{k}|\lambda'\mathbf{k}\rangle\langle\lambda\mathbf{k}| - |\lambda'\mathbf{k}\rangle\langle\lambda\mathbf{k}|v\mathbf{k}\rangle\langle c\mathbf{k}|\right)\Big]$$

Equations of motion for density matrix elements

$$\rho_{cv}^{int}(\mathbf{k},t) = \langle c\mathbf{k}|\rho_{\mathbf{k}}^{int}(t)|v\mathbf{k}\rangle$$

$$\frac{d}{dt}\rho_{cv}^{int}(\mathbf{k},t) = \frac{i}{\hbar}d_{cv}\mathcal{E}(t)e^{i(\epsilon_{c,\mathbf{k}}-\epsilon_{v,\mathbf{k}})t}[\rho_{vv}(\mathbf{k},t) - \rho_{cc}(\mathbf{k},t)]$$

$$\frac{d}{dt}\rho_{cc}(\mathbf{k},t) = \frac{i}{\hbar}\mathcal{E}(t)\left[d_{cv}e^{i(\epsilon_{c,\mathbf{k}}-\epsilon_{v,\mathbf{k}})t}\rho_{vc}^{int}(\mathbf{k},t) - c.c.\right]$$

$$\frac{d}{dt}\rho_{vv}(\mathbf{k},t) = \frac{i}{\hbar}\mathcal{E}(t)\left[d_{cv}^{*}e^{i(\epsilon_{v,\mathbf{k}}-\epsilon_{c,\mathbf{k}})t}\rho_{cv}^{int}(\mathbf{k},t) - c.c.\right] = -\frac{d}{dt}\rho_{cc}(\mathbf{k},t)$$

Two important limiting cases of the noninteracting system:
coherent optical interband transitions;
quasi-equilibrium electron-hole plasma

Coherent regime

Electromagnetic field: $\mathcal{E}(t) = \frac{E_0}{2}(e^{i\omega t} + e^{-i\omega t})$ $\rho_{cv}(\mathbf{k},t) = \rho_{cv}^{int}(\mathbf{k},t)e^{-i(\epsilon_{c,\mathbf{k}}-\epsilon_{v,\mathbf{k}})t}$

Interband equation:

$$\left(\frac{d}{dt} + i\nu_{\mathbf{k}}\right)\rho_{cv}(\mathbf{k},t)e^{i\omega t} = -\frac{i\omega_R}{2}[\rho_{cc}(\mathbf{k},t) - \rho_{vv}(\mathbf{k},t)]$$

$$\frac{d}{dt}\rho_{cc}(\mathbf{k},t) = -\frac{i}{2}\omega_R[\rho_{cv}(\mathbf{k},t)e^{i\omega t} - \rho_{vc}(\mathbf{k},t)e^{-i\omega t}] = -\frac{d}{dt}\rho_{vv}(\mathbf{k},t)$$

with detuning $\nu_{\mathbf{k}} = \epsilon_{c,\mathbf{k}} - \epsilon_{v,\mathbf{k}} - \omega$ and Rabi frequency $\omega_R = \frac{d_{cv}E_0}{\hbar}$

Define Bloch vector:

$$U_1(\mathbf{k},t) = 2\ \mathrm{Re}[\rho_{cv}(\mathbf{k},t)e^{i\omega t}]$$
$$U_2(\mathbf{k},t) = 2\ \mathrm{Im}[\rho_{cv}(\mathbf{k},t)e^{i\omega t}]$$
$$U_3(\mathbf{k},t) = [\rho_{cc}(\mathbf{k},t) - \rho_{vv}(\mathbf{k},t)]$$

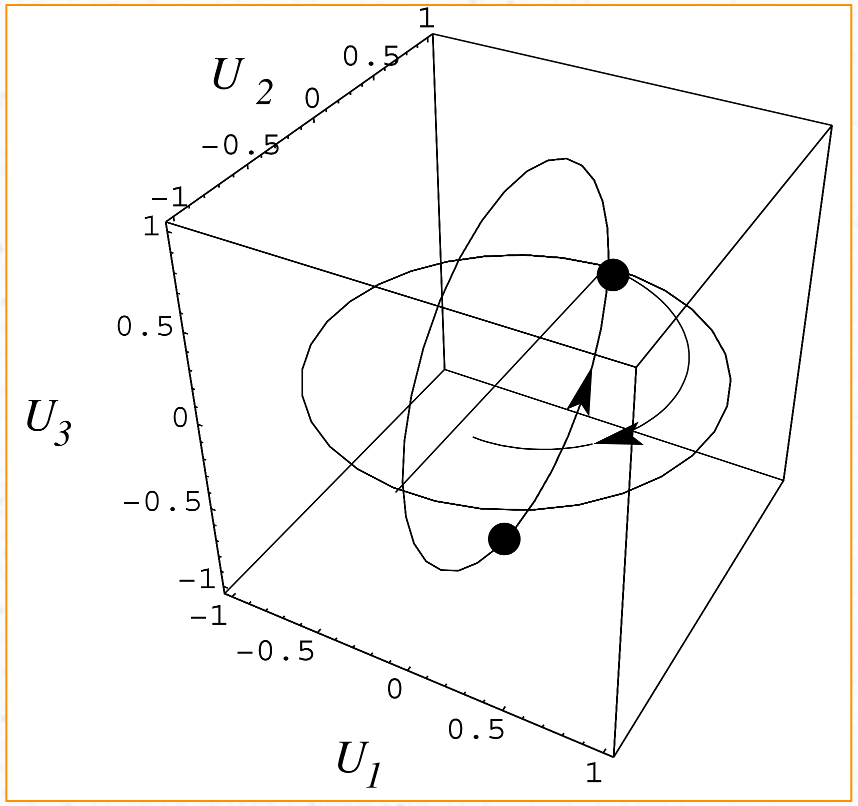
Coherent optical Bloch equations:

$$\frac{d}{dt}U_1(\mathbf{k},t) = \nu_{\mathbf{k}}U_2(\mathbf{k},t)$$
$$\frac{d}{dt}U_2(\mathbf{k},t) = -\nu_{\mathbf{k}}U_1(\mathbf{k},t) - \omega_R U_3(\mathbf{k},t)$$
$$\frac{d}{dt}U_3(\mathbf{k},t) = \omega_R U_2(\mathbf{k},t)\ .$$

$$\Longrightarrow \quad \frac{d}{dt}\mathbf{U}(\mathbf{k},t) = \mathbf{\Omega}\times\mathbf{U}(\mathbf{k},t)\ ,\quad \mathbf{\Omega} = \omega_R\,\mathbf{e}_1 - \nu_{\mathbf{k}}\,\mathbf{e}_3$$

Optical Bloch equations with relaxation

$$\frac{d}{dt}U_1(\mathbf{k},t) = -\frac{U_1(\mathbf{k},t)}{T_2} + \nu_{\mathbf{k}}\,U_2(\mathbf{k},t)$$
$$\frac{d}{dt}U_2(\mathbf{k},t) = -\frac{U_2(\mathbf{k},t)}{T_2} - \nu_{\mathbf{k}}\,U_1(\mathbf{k},t) - \omega_R\,U_3(\mathbf{k},t)$$
$$\frac{d}{dt}U_3(\mathbf{k},t) = -\frac{U_3(\mathbf{k},t)+1}{T_1} + \omega_R\,U_2(\mathbf{k},t) \ .$$



Free induction decay $\omega_R = 0$:

$$\mathbf{U}(\mathbf{k},t=0) = U_1(\mathbf{k},t=0)\ \mathbf{e}_1 + U_2(\mathbf{k},t=0)\ \mathbf{e}_2$$

$$\frac{d}{dt}U_1(\mathbf{k},t) = -\frac{U_1(\mathbf{k},t)}{T_2} + \nu_{\mathbf{k}}\,U_2(\mathbf{k},t)$$
$$\frac{d}{dt}U_2(\mathbf{k},t) = -\frac{U_2(\mathbf{k},t)}{T_2} - \nu_{\mathbf{k}}\,U_1(\mathbf{k},t)$$

$$\begin{pmatrix} U_1(\mathbf{k},t) \\ U_2(\mathbf{k},t) \end{pmatrix} = \begin{pmatrix} \cos(\nu_{\mathbf{k}}t) & \sin(\nu_{\mathbf{k}}t) \\ -\sin(\nu_{\mathbf{k}}t) & \cos(\nu_{\mathbf{k}}t) \end{pmatrix} \begin{pmatrix} U_1(\mathbf{k},t) \\ U_2(\mathbf{k},t) \end{pmatrix} e^{-t/T_2}$$

Photon echo intensity: $[e^{-2\tau/T_2}]^2 = e^{-4\tau/T_2}$

Free carrier absorption in quasi-equilibrium regime

Thermal equilibrium distribution for electrons:

$$\rho^0_{\lambda\lambda} = \frac{1}{e^{(\epsilon_{\lambda,\mathbf{k}}-\mu_\lambda)\beta}+1} \equiv f_{\lambda,k} \qquad \sum_{\mathbf{k}} f_{\lambda,k} = N_\lambda \rightarrow \mu_\lambda = \mu_\lambda(N_\lambda, T)$$

$$\rho^{int}_{cv}(\mathbf{k},t) = \int \frac{d\omega}{2\pi} \frac{d_{cv}\mathcal{E}(\omega)e^{i(\epsilon_{c,\mathbf{k}}-\epsilon_{v,\mathbf{k}}-\omega)t}}{\hbar(\epsilon_{c,\mathbf{k}}-\epsilon_{v,\mathbf{k}}-\omega-i\gamma)}(f_{v,k}-f_{c,k})$$

Optical polarisation:

$$\mathcal{P}(t) = \mathrm{tr}[\rho(t)d] = \mathrm{tr}[\rho^{int}(t)d^{int}(t)] = \frac{1}{L^3}\sum_{\mathbf{k}}[\rho^{int}_{cv}(\mathbf{k},t)d^{int}_{vc}(\mathbf{k},t) + \rho^{int}_{vc}(\mathbf{k},t)d^{int}_{cv}(\mathbf{k},t)]$$

$$= \frac{1}{L^3}\sum_{\mathbf{k}}\int \frac{d\omega}{2\pi} \frac{|d_{cv}|^2(f_{v,k}-f_{c,k})}{\hbar(\epsilon_{c,\mathbf{k}}-\epsilon_{v,\mathbf{k}}-\omega-i\gamma)}\mathcal{E}(\omega)e^{-i\omega t} + \mathrm{c.c.}$$

Optical susceptibility:

$$\chi(\omega) = \mathcal{P}(\omega)/\mathcal{E}(\omega) \qquad \mathcal{P}(t) = \int \frac{d\omega}{2\pi}\mathcal{P}(\omega)e^{-i\omega t}$$

Absorption spectrum

Optical susceptibility for free carriers:

$$\chi(\omega) = -\sum_{\mathbf{k}} \frac{|d_{cv}|^2}{L^3}(f_{v,k} - f_{c,k}) \left[\frac{1}{\hbar(\epsilon_{v,\mathbf{k}} - \epsilon_{c,\mathbf{k}} + \omega + i\gamma)} - \frac{1}{\hbar(\epsilon_{c,\mathbf{k}} - \epsilon_{v,\mathbf{k}} + \omega + i\gamma)}\right]$$

Absorption spectrum:

$$\alpha(\omega) = \frac{4\pi\omega}{n_b c}\chi''(\omega) = \frac{4\pi^2\omega}{L^3 n_b c}\sum_{\mathbf{k}} |d_{cv}|^2 (f_{v,k} - f_{c,k})\,\delta\left[\hbar(\epsilon_{v,\mathbf{k}} - \epsilon_{c,\mathbf{k}} + \omega)\right]$$

For different dimensionalities D:

$$f_{h,k} = 1 - f_{v,k} \qquad \hbar(\epsilon_{c,k} - \epsilon_{v,k}) = \hbar(\epsilon_{e,k} + \epsilon_{h,k}) = \frac{\hbar^2 k^2}{2m_r} + E_g$$

$$\alpha(\omega) = \frac{8\pi^2\omega|d_{cv}|^2}{n_b c L_c^{3-D}} \frac{1}{(2\pi)^D}\Omega_D S_D(\omega)$$

$$S_D(\omega) = \int_0^\infty dk\, k^{D-1}\delta\left(\frac{\hbar^2 k^2}{2m_r} + E_g + E_0^{(D)} - \hbar\omega\right)(1 - f_{e,k} - f_{h,k})$$

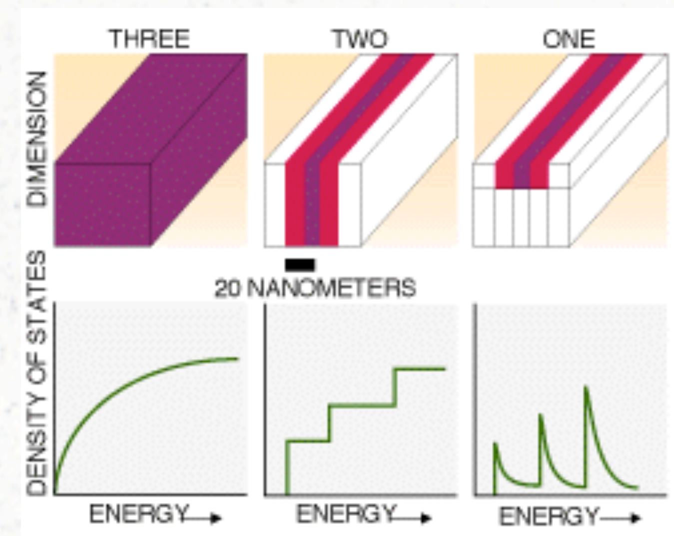
$$E_0^{(D)} = \frac{\hbar^2}{2m_r}\left(\frac{\pi}{L_c}\right)^2 (3 - D)$$

Absorption coefficient

Density of states for dimensionality D:

$$S_D(\omega) = \frac{1}{2}\left(\frac{2m_r}{\hbar^2}\right)^{D/2}\int_0^\infty dx\, x^{(D-2)/2}\,\delta(x + E_g + E_0^{(D)} - \hbar\omega) \times [1 - f_e(x) - f_h(x)]$$

$$\frac{\hbar^2 k^2}{2m_r} = x$$



$$S_D(\omega) = \frac{1}{2}\left(\frac{2m_r}{\hbar^2}\right)^{D/2}(\hbar\omega - E_g - E_0^{(D)})^{(D-2)/2} \times\Theta(\hbar\omega - E_g - E_0^{(D)})A(\omega)$$

Absorption coefficient for free carriers:

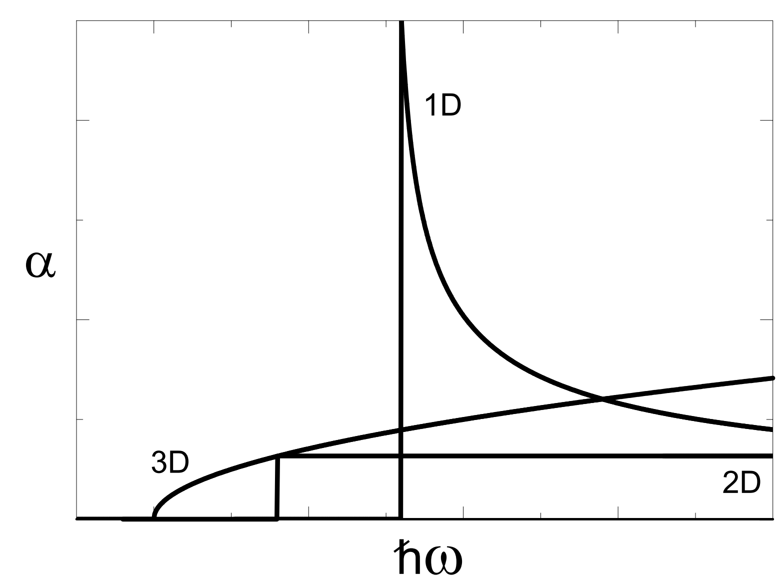
$$\boldsymbol{\alpha(\omega) = \alpha_0^D \frac{\hbar\omega}{E_0}\left(\frac{\hbar\omega - E_g - E_0^{(D)}}{E_0}\right)^{\frac{D-2}{2}}\Theta(\hbar\omega - E_g - E_0^{(D)})A(\omega)}$$

$$3\text{D} \sim \sqrt{\hbar\omega - E_g}$$

$$2\text{D} \sim \Theta(E_g + E_0^{(2)})$$

$$1\text{D} \sim 1/\sqrt{\hbar\omega - E_g - E_0^{(1)}}$$

Unexcited material:



Negative optical absorption

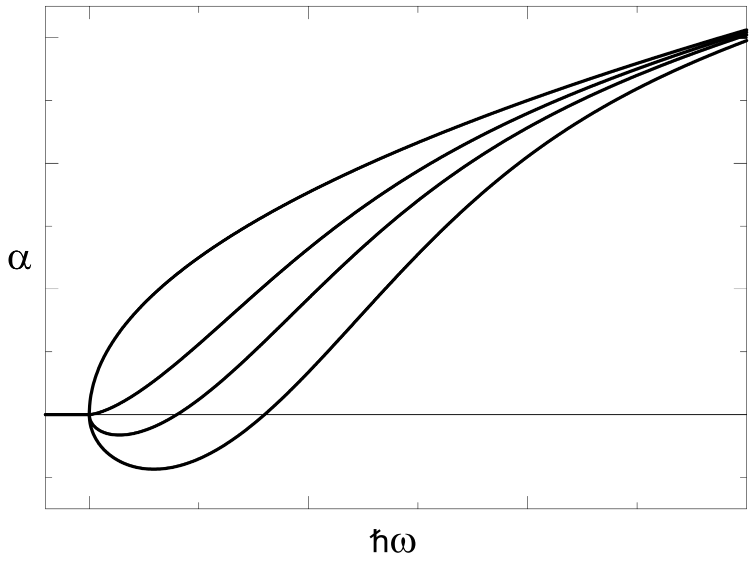
Band filling factor:

$$A(\omega) = \underbrace{\left[(1 - f_e(\omega))(1 - f_h(\omega)) + f_e(\omega) f_h(\omega)\right]}_{0 \leq f_{e/h} \leq 1 \;\Rightarrow\; 0.5 < \ldots < 1} \underbrace{\tanh\left[\frac{\beta}{2}(\hbar\omega - E_g - \mu)\right]}_{<0}$$

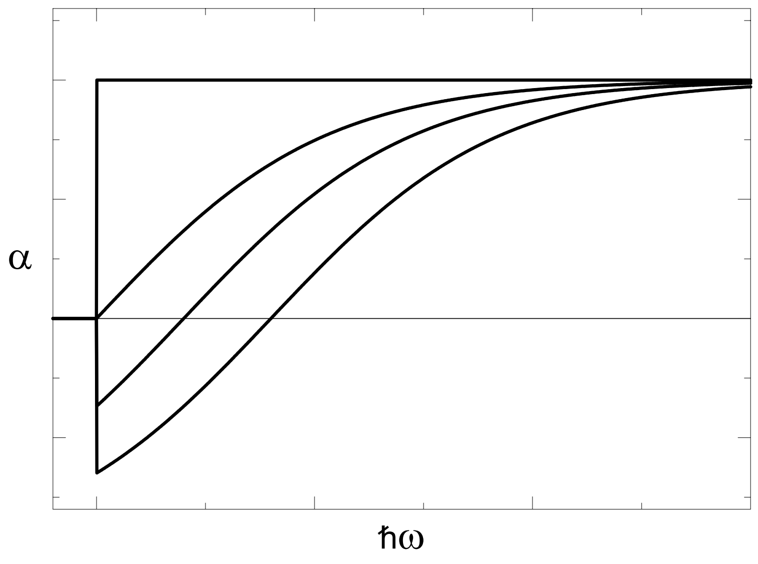
if $\mu > 0$ and $E_g < \hbar\omega < E_g + \mu$

Absorption/gain spectra for D-dimensional semiconductors:
(carrier density grows from top to bottom)

3D



2D



1D